Superconducting properties of $[BaCuO_x]_2 / [CaCuO_2]_n$ artificial structures with ultrathick $CaCuO_2$ blocks

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Abstract

The electrical transport properties of $[BaCuO_x]_2/[CaCuO_2]_n$ ($CBCCO - 2 \times n$) underdoped high temperature superconducting superlattices grown by Pulsed Laser Deposition have been investigated. Starting from the optimally doped $CBCCO - 2 \times 2$ superlattice, having three CuO_2 planes and T_c around 80 K, we have systematically increased the number n up to 15 moving toward the underdoped region and hence decreasing T_c. For $n > 11$ the artificial structures are no longer superconducting, as expected, for a uniformly distributed charge carriers density inside the conducting block layer. The sheet resistance of such artificial structures ($n \approx 11$) turns out to be quite temperature independent and close to the 2D quantum resistance 26 $k\Omega$. A further increase of the number of CuO_2 planes results in an insulator-type dependence of $R(T)$ in the wide range of temperatures from room temperature to 1 K. The value of the sheet resistance separating the Superconducting and the Insulating regimes supports the fermionic scenario of the Superconductor-Insulator transition in these systems.

A. Introduction

Recently a large effort has been devoted to the layer-by-layer growth of high temperature superconducting cuprate (HTS) superlattices. Among them, particularly interesting are the HTS superlattices $[BaCuO_x]_m/[CaCuO_2]_n$ ($CBCCO - m \times n$), with $m \simeq 2$ [1]. These artificial structures are grown by Pulsed Laser Deposition (PLD) stacking in sequence non-superconducting individual layers of $BaCuO_x$ and $CaCuO_2$ In the $CBCCO - m \times n$ unit cell, the superconducting block, named Infinite Layer (IL) block, consists of n epitaxial layers of $CaCuO_2$, while the Charge Reservoir (CR) block is made of m epitaxial layers of $BaCuO_x$ [2]. In the case of $[BaCuO_x]_2/[CaCuO_2]_2$ (2×2 superlattices) grown at relatively high molecular oxygen pressure ($\simeq 1$ $mbar$) and at a temperature of about 650 $^\circ C$, it has

been shown that the doping is nearly optimal ($p \simeq 0.18 - 0.19$ holes/CuO_2 plane) and $T_c \simeq 80\ K$ [3,4].

In our previous work on the $CBCCO - 2 \times n$ system, we investigated both the dependence of the critical temperature on n (for n ranging from 1 to 6) and the influence of the interfacial structural disorder, typical for these artificial structures, on the electrical transport properties [4,5]. It was found that the critical temperature reaches the maximum value of about 80 K for n between 2 and 3. It decreases both for n larger than 3 and smaller than 2. Such an effect was explained on the basis of the structural disorder (especially effective for small n) and of the variation of the effective carriers concentration with n. Furthermore it was demonstrated that some peculiar features of the temperature dependence of the resistivity (its relatively high residual value, its pronounced rounding above T_c and its saturation above room temperature) can be well explained by taking into account the role of the interfacial structural disorder that arises at the blocks interfaces [4]. Following this considerations, we believe that the role of the structural disorder is strongly reduced for the superlattices with thick IL blocks, negligible for the thickest ones.

In this paper we report on the investigation of the electrical transport properties of $[BaCuO_x]_2/[CaCuO_2]_n$ superlattices having ultrathick IL layers. For this purpose we have grown many $2 \times n$ superlattices having the same CR block, consisting of 2 epitaxial layers of $BaCuO_x$, but different superconducting blocks containing $(n+1)$ CuO_2 planes [5]. We varied the number of $CaCuO_2$ layers per cell from 2 up to 15, keeping constant all the growth parameters and the thickness of the CR block of the superlattices. In this way we decreased the average doping level per CuO_2 plane inside the superconducting block, starting from the optimally doped $CBCCO - 2 \times 2$ superlattice with T_c around 80 K. An important experimental finding was that for $n > 11$ (corresponding to the critical value $p \simeq 0.04$ holes/CuO_2 plane) the critical temperature goes to zero and the resistivity versus temperature follows closely the behavior predicted by the variable range hopping mechanism. Such a result indicates that the distribution of the charge carriers in the IL block is probably quite uniform without sizeable confinement at the interfaces between the IL block and the

CR parent block. On the contrary, in the case of the charge localization at the CR/IL interfaces, one would expect a saturation of the critical temperature value for a sufficiently thick IL block (as in the case of twin-planes superconductivity [6]).

Therefore, starting from the optimally doped $CBCCO-2\times 2$ superlattice we have systematically increased the number n of CuO_2 planes per cell, moving toward the underdoped part of the phase diagram and hence decreasing T_c. In this way the doping level (per CuO_2 plane) becomes smaller and smaller, until a critical value of the IL block thickness is reached at which superconductivity disappears. We show that, for the 2×11 superlattice, the superconducting critical temperature tends to zero while $R_{\Box}$ approaches the value of 25.8 $k\Omega$ close to the universal quantum resistance for the $2D$ Superconductor-Insulator (S-I) transition [7]. For $n>11$ the $R(T)$ behavior was found to be of the insulator-type. The results obtained suggest that the S-I transition, occurring at $n\simeq 11$, can be explained within the fermionic scenario, where the insulating state of $2D$ electrons system is reached when the charge carriers density is so low that $k_F l\simeq 1$.

B. Experimental results

Artificial structures with n ranging from 2 to 15 have been grown by PLD on $SrTiO_3(001)$ substrates. Two targets, with $CaCuO_2$ and $BaCuO_2$ nominal composition respectively, mounted on a multitarget system, were used. The oxygen pressure and the substrate temperature, during the deposition, were 1 $mbar$ and 650 $^{\circ}C$ respectively. Further details on the growth procedure are given in Ref. [2]. Films have thickness ranging from 600 to 1000 Å. The samples crystallographic structure were characterized by X-ray diffraction (XRD) analysis.

The unit cells thickness Λ varies between 15.2 Å (2×2 superlattices) and 56.8 Å (2×15 superlattices). The thickness of the IL block has been varied increasing the number of laser shots on the $CaCuO_2$ target. All films were found to have a very small value of the mosaic spread ($\simeq 0.08^{\circ}$), close to the substrate one ($\simeq 0.06^{\circ}$). In Fig.2 the $SL_0(002)$ rocking curve

of the superconducting 2×2 superlattice (a) and the $SL_{+1}(001)$ of the insulating 1.9×14 superlattice (b) are shown, the superlattices peaks, $SL_{\pm N}$ $(00l)$, are the N^{th} order satellite peaks of the substrate $00l$ reflections, the $zero^{th}$ order being the average structure peak of the artificial structure.

The XRD spectra of the $[BaCuO_x]_2/[CaCuO_2]_n$ superlattices are shown by empty dots in Fig.1 for four different values of n. The effective thickness of both the CR and IL block were deduced by a kinematical simulation of the diffraction spectra (full lines in Fig.1). This simulation is achieved considering that a two dimensional layer-by-layer growth occurs and that layers of mixed composition are corrugated to adjust the internal stresses. A random disorder is added to take into account the experimental dispersion in the amount of deposited material in each iteration [8]. In our simulations the random thickness fluctuations for each deposited layer is found to be indipendent from n. Furthermore a small decrease in the deposition rate during the growth has been considered in order to reproduce the drastic suppression of the $SL_{\pm 3}$ and $SL_{\pm 4}$ peaks. Such an effect is expected due to the progressive damage of the target surface during the growth. The agreement between the experimental data and the simulated spectra is very good. All the major features of the diffraction spectra are reproduced by the simulations.

Preliminary asymmetrical reflections XRD measurements, carried out at the European Synchrotron Radiation Facility, have shown that the in-plane crystallographic directions a, b of the superlattices are aligned with those of the $SrTiO_3(001)$ substrates with an uncertainty of 0.15° [9].

Resistivity measurements were performed by the standard four-probe dc technique. Contacts were made by silver epoxy directly on the substrate before the film deposition, in order to avoid any chemical reaction between the superlattices and the solvent utilized in the silver epoxy. The probe current density was about 100 A/cm^2. The temperature was varied at a typical rate of $1K/min$, with an accuracy of 10 mK. The sheet resistance $R_\square$ was calculated, for each superlattice, assuming the distance between two $2D$ successive conducting block

layers to be equal to the specific modulation length Λ : $R_{\square} = \frac{\rho_{ab}}{\Lambda}$.

The behavior of sheet resistance versus temperature is shown in Fig.3a for several superlattices with different n values. By increasing the number of CuO_2 planes in the IL block, the following striking features can be noticed:

a). T_c goes smoothly to zero. For the 2×11 superlattices (having 12 CuO_2 planes in the IL block) the critical temperature (zero resistance point) is 1 K, while the 2×14 and 2×15 superlattices show an insulating behavior with no trace of superconductivity above 1 K.

b). The sheet resistance $R_{\square}$ becomes larger. All the $CBCCO$ films with $n > 8$ show an upturn of resistivity with a negative $d\rho/dT$ (see the Fig.3b) at low temperature above the superconducting transition.

c). The crossover between metallic (positive $d\rho/dT$) and insulating (negative $d\rho/dT$) low temperature behavior occurs for $n > 8$. Films with the thickest IL block have sheet resistance higher than 30 $k\Omega$.

Curves belonging to *superconducting* and *insulating* phases could be ideally separated by a separatrix lying near the 2×11 superlattice $R(T)$ dependence. This ideal separatrix curve has a sheet resistance of the order of quantum $2D$ resistance ($\simeq$ 26 $k\Omega$) (see the detailed discussion of the properties of this critical region in [10]). As one can see from Fig. 5 films with $n > 11$ are not superconducting: this behavior was expected because we can estimate a doping level $p < 0.04$ holes/CuO_2 plane, below the critical value for superconductivity in many HTS cuprates [11,12]. All films with 15 or more CuO_2 planes in the IL block showed the variable range hopping (VRH) behavior typical of 2D insulators. This point is stressed in the inset of Fig.5, where the $ln(R)$ is reported as a function of $(1/T)^{1/3}$. The excellent linear behavior of the experimental data can be noted. The above scenario is also enforced by the inset of Fig.4: here a clear inverse correlation between the critical temperature and the sheet resistance can be noted: T_c goes to zero as $R_{\square}$ reaches $25 - 30$ $k\Omega$.

C. Discussion

The first important finding concerns the charge carriers distribution over the IL block. The disappearance of superconductivity for the samples with thick enough IL blocks indicates that the carriers are distributed quite homogeneously along the $c-$axis and that their density decreases increasing n. Assuming $\rho(n) = \rho_{opt}\ \frac{3}{n+1}$, with $\rho_{opt} \simeq 0.18$, one can estimate the critical doping value $\rho(n = 11) \simeq 0.045$ at which the phenomenon of superconductivity disappears. It is clear that this value is very close to the critical doping of the HTS materials ($\simeq 0.04$). [11,12]

Another interesting aspect of our investigation is the new way to drive the system through the S-I transition: namely, leaving unchanged the overall number of carriers provided by the CR block, we have varied the effective carriers density, varying the number of "conducting" CuO_2 planes in the IL block.

The S-I transition has been widely studied during the last years in a variety of disordered and underdoped systems: in granular thin metallic film such as Bi, Sn, Pb, Al [13], and in high-T_c cuprates such as irradiated $Y-Ba-Cu-O$ and $Bi-Sr-Ca-Cu-O$ [7,14], $Zn-$doped $La-Ba-Ca-Cu-O$ [15], $Y-$doped $Bi-Sr-Ca-Cu-O$ [16], $Ce-$doped $Nd-Cu-O$ [17], oxygen-deficient $Y-Ba-Cu-O$ single crystals [18], $Pr-$doped or $Zn-$doped $Y-Ba-Cu-O$ [19,20], ultrathin $Dy-Ba-Cu-O$ films [21]. In this context it is evident that our superlattices are not the simplest objects for the study of such fundamental phenomenon like the S-I transition. Nevertheless they show some advantages relative to other systems. Namely, due to the complexity of the unit cells of the HTS materials, an evident ambiguity exists concerning the definition of the effective thickness t of the conducting layer necessary to calculate $R_{\Box}$. It can be seen that, in the case of many HTS compounds, the specific choice of the thickness of the conducting layer (either the whole unit cell that is the CR plus the IL block, or the IL block alone, or the single CuO_2 plane) can result in a large uncertainty in the $R_{\Box}$ value. For instance, in the case of $Bi_2Sr_2Y_xCa_{1-x}Cu_2O_8$ compound [16] the thickness t of the conducting layer has been

chosen as the $CuO_2/Ca/CuO_2$ layers distance (namely the half of $c-$axis lattice constant, $t \simeq 15$ Å), instead in Ref. [18] t is equal to the $c-$axis lattice constant of $Y_1Ba_2Cu_3O_{7-x}$ ($t \simeq 11.8$ Å). On the other hand, in the case of $Nd_{2-x}Ce_xCu_1O_4$ single crystal [17], t is the lattice spacing between CuO_2 layers ($t \simeq 6$ Å). This ambiguity turns out to be crucial for the possibility of judging which type of scenario of the 2D S-I transition (bosonic or fermionic) is realized in practice [22].

The first mechanism (bosonic), is based on the duality hypothesis. In fact, both classical and quantum fluctuations reduce the superfluid density ρ_s and therefore, suppress the Berezinski-Thouless-Kosterlitz transition temperature $T_c^{\rm BKT}$. The magnitude of this suppression is determined by the Ginzburg-Levanyuk number Gi_2 which turns out to be of the order of 1 when the dimensionless conductance $g = \dfrac{\sigma}{e^2/h} \to g_c \sim 1$. As a result, at this level of disorder, the superfluid density, simultaneously with $T_c^{\rm BKT}$, becomes zero. In the vicinity of this critical carriers concentration is $T_c^{\rm BKT} \ll T_{c(0)}$ (where $T_{c(0)}$ is the mean field critical temperature) and in the temperature range $T_c^{\rm BKT} \ll T \ll T_{c(0)}$, the conductivity is realized by fluctuating Cooper pairs. The problem of quantum liquid motion near the quantum phase transition can be approached from a second view point. It can be said that, with the increase of Gi_2, the role of quantum fluctuations grows and that fluctuating vortices, carrying the magnetic flux quantum $\Phi_0 = h/2e$, are generated. The duality hypothesis [23] assumes that, at the critical point, the pair and vortex liquid density flows are equal, giving, for the conductivity the universal value, at the critical point: $\sigma_c = \frac{2e}{\Phi_0} = \frac{4e^2}{h}$.

The second, fermionic, mechanism of the S-I transition is related to the renormalization of the inter-electron interaction in the Cooper channel by the long-range Coulomb repulsion, specific to dirty $2D$ superconductors [24,25]. As long as the correction to the unrenormalized BCS transition temperature $T_{c(0)}$ is still small, it is found that: $T_c = T_{c(0)}\left(1 - \dfrac{1}{12\pi^2 g}\ln^3\dfrac{\hbar}{k_B T_{c(0)}\tau}\right)$. The suppression of T_c down to zero in this case may occur even for $g \gg 1$. The renormalization group analysis gives [26], for the critical value of conductance: $g_c = \left(\frac{1}{2\pi}\ln\frac{\hbar}{k_B T_{c(0)}\tau}\right)^2$. At high degree of disorder (or low level of doping), $T_{c(0)}\tau$

is small enough, so that $\ln \frac{\hbar}{k_B T_{c(0)} \tau} > 5$: the fermionic mechanism of the critical temperature suppression turns out to be of primary importance. In this case $g_c > 2/\pi$: the boson mechanism turns out to be insignificant. On the contrary, if $\ln \frac{\hbar}{k_B T_{c(0)} \tau} < 4$, the correction for T_c is small even for $g_c = 2/\pi$, so the boson mechanism becomes of primary importance. The typical experimental values of g_c are in the region $g_c \sim 1$, and do not differ dramatically from the predictions of the boson duality assumption $g_c = \frac{2}{\pi}$ (by estimating for the 2×2 superlattices [5] $\tau \simeq 2 \times 10^{-15}$ *sec*, $T_{c(0)} = 80$ *K*, g_c turns out to be quite larger than $\frac{2}{\pi}$). This is why the precise evaluation of the experimental value of the sheet resistance is so important. In the case of our samples, with ultrathick IL blocks, the ambiguity in determining the conducting layer thickness is minimal. In fact, this ambiguity is of the order of the CR block thickness in comparison with that one of the much thicker IL block. Therefore the value of $R_{\Box} \simeq 25 - 30\ k\Omega$, found according to our experiments, is highly reliable and allows us to conclude that the observed S-I transition has a fermionic character. An additional argument in favor of this conclusion can be found in the logarithmic growth of the resistance in the metal phase of the $2 \times (9 \div 11)$ samples (see Fig.5) which is coherent with the weak localization theory.

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FIGURES

FIG. 1. XRD spectra of four $m \times n$ superlattices with ultrathick IL blocks (open circles) and the corresponding simulated spectra (full line) obtained by the procedure described in Ref. [8]. The $SrTiO_3(001)$ peaks are labeled by the asterisks.

FIG. 2. Rocking curves: a) $SL_0(002)$ peak of the 2×2 superconducting superlattice. b) $SL_{+1}(001)$ peak of the 1.9×14 insulating superlattice. The full width at half maximum (FWHM) were estimated by Lorentzian curve fits.

FIG. 3. a) Sheet resistance versus temperature of $CBCCO - m \times n$ superlattices in which m is nearly 2. b) First derivative of the resistance, normalized to its 300 K value, of the same superlattices.

FIG. 4. Critical temperature (zero resistance point) versus the number of CuO_2 planes in the IL block. The vertical dotted line ideally divides the overdoped from the underdoped region. Inset: critical temperature versus the sheet resistance. Here the sheet resistance values were obtained as the $T = 0$ K linear extrapolation of ρ_{ab}/Λ, where Λ is the thickness of the unit cell.

FIG. 5. Normalized resistance versus T (20 $K < T < 300$ K) in semilogarithmic scale of two ultrathick superconducting superlattices. Inset: $2D$ VRH behavior of the thickest structures.